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Results of Gamma-Ray Measurements from a Recent Demonstration for Russian Technical Experts

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Results of gamma-ray measurements from a recent demonstration for Russian technical experts[†]

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Abstract

In August 2001, a group of U.S. technical experts demonstrated an Attribute Measurement System with an Information Barrier (AMSIB) for a delegation of Russian technical experts. The purpose of the demonstration was to show that attributes of a classified plutonium item of potential interest to arms control and nonproliferation transparency regimes could be ascertained without releasing any sensitive information. For this demonstration, both gamma-ray and neutron attributes were determined. We consider only the gamma-ray attributes here. The specific plutonium attributes measured were the isotopic ratio of ^{240}Pu to ^{239}Pu, the "age" of the plutonium (time elapsed since the most recent chemical purification of the plutonium), and the absence of plutonium oxide in the item's storage container. In this paper, we briefly review the technologies employed for the attribute measurements used in the gamma-ray portion of the demonstration, concentrating on the results of the test measurements of the isotopic and age attributes made on unclassified items.

Introduction

Last year, we presented the methods we used to determine various plutonium attributes from gamma-ray signatures of plutonium items at the Fissile Material Transparency Technology Demonstration (FMTTD).[1] These were—

1. Presence of plutonium
2. Plutonium age (time elapsed since the last americium separation)
3. Presence of weapons-quality plutonium (defined as ^{240}Pu/^{239}Pu < 0.1)
4. Absence of plutonium oxide

The demonstration simulated part of a hypothetical transparency regime in which plutonium items would be accepted into the regime provided they exhibited satisfactory results from attribute measurements. Because the actual values of the results could be classified, they were not revealed but compared within a computer with unclassified thresholds. If the results for all four measurements fell appropriately above (or below for some attributes) their threshold values, the item "passed" and was accepted into the inspection regime.

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Three codes for determining the plutonium gamma-ray attributes—called Pu-300, Pu-600, and Pu-900—were developed or upgraded for the FMTTD. Pu-300 obtains the time elapsed since the last ^{241}Am chemical separation. This is based on determining the ^{241}Am to ^{241}Pu ratio in the energy region from 330–350 keV, hence the name Pu-300. Pu-600 determines the isotopic ratio of ^{240}Pu to ^{239}Pu to determine that the plutonium is weapons-quality. This analysis is carried out in the energy region of 630–670 keV. Pu-900 determines the presence of PuO_2 by searching for a peak at 870.7 keV. If this peak is present, it is consistent with the presence of PuO_2 in the sample. The peak arises from the de-excitation of the first excited state of ^{17}O.

These codes are part of a system that includes software and hardware. The hardware was assembled mostly from commercial components. Gamma radiation is measured with HPGe detectors (Fig. 1). The Pu-300 and Pu-600 methods share one detector and their measurements are made serially. This is a detector of 50% relative efficiency and of n-type to minimize neutron damage to the crystal. The signals from the output of the detector preamplifer are sent to two destinations. The first output goes to the control electronics for a mechanical tungsten iris whose aperture is automatically adjusted to maintain system dead time at a relatively constant value. The iris was concealed in a RF-shielded enclosure so that no information can be inferred about the strength of the source by observing the iris aperture. By keeping the counting time and detector-to-source distance constant and the iris aperture concealed, no source strength information can be inferred.

The second output from the HPGe detector preamplifier is fed to a Canberra InSpector™ data-acquisition system. This instrument combines the functions of a bias supply, spectroscopy amplifier, and multichannel analyzer. We controlled the InSpector for automated operation using a program written expressly for this purpose that runs under the MSDOS™ operating system. This control program relies on the Canberra MSDOS libraries. The control-and-analysis computer was integrated using commercial PC104 bus components and included a single board computer. The computer is booted from a PROM and otherwise contains only volatile memory. In addition to the MSDOS operating system, the PROM contains the control and analysis software for the system. The system is fully automated and the only action that needs to be taken by the operator is to press a single button to initiate the analysis.

Pu-300—Plutonium age

During the months that led up to the FMTTD, we measured the age of a variety of plutonium samples. These samples ranged in age from several months to thirty years. To shake out the bugs in the Pu-300 method, we had to resolve two major issues.

The items had to be viewed through the side of an improvised neutron multiplicity counter. We found that differential attenuation in the high-density polyethylene of the counter proved significant for Pu-300. There was about a 7% differential attenuation of the gamma rays at 332- and 345-keV that, without correction, caused the algorithm to be quite unstable. This instability was remedied by applying a correction to the algorithm based on the measured differential attenuation.

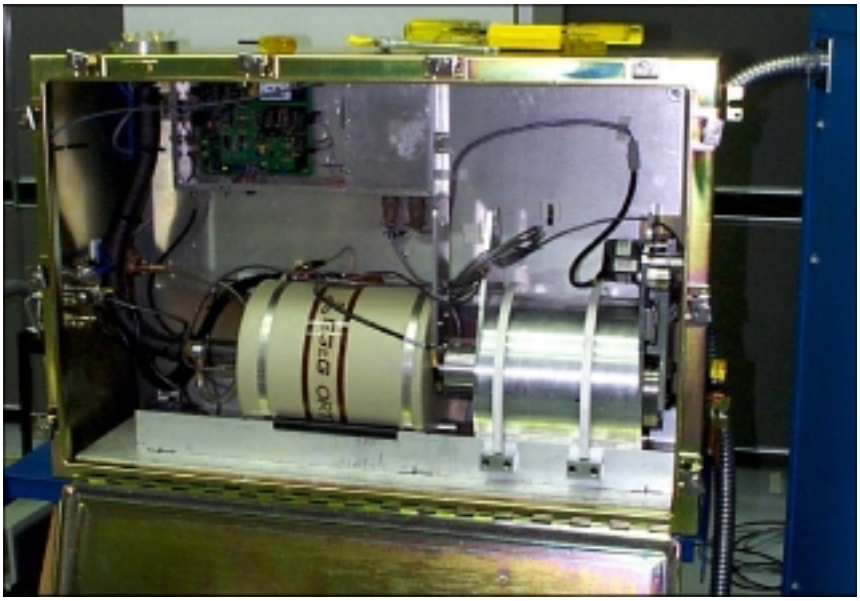

Figure 1. The Pu-300 and Pu-600 detector system with its RF enclosure, an information barrier element, shown open. A 2.0-cm machinable tungsten shield surrounds the commercial detector cryostat. To the right of the shield, viewed edge-on, is the autonomous tungsten iris. In the upper left-hand side of the enclosure is the iris control circuitry

The thick high-density polyethylene in the side of the multiplicity counter provided not only significant differential attenuation but total attenuation as well—absorbing about 75% of the gamma rays in our energy region. The Pu-300 method is intended to obtain results of adequate accuracy and precision from hundreds of grams of material after counting for 15 minutes. Two items used in the demonstration were unclassified sources with approximately known attributes. One item, five fuel plates from the Zero Power Physics Reactor (ZPPR), was chosen for its convenience rather than for its appropriateness as an example for an actual transparency regime. This item had a mass of only 122 grams and a flat-plate geometry that, when viewed end on, produced an unacceptably low count rate. We solved this problem by determining an optimal counting geometry for the source measurement.

The test results for the ZPPR-plate age measurements are plotted in Fig. 2. The average measured age from these 59 measurements is 33.9 ± 1.6 years. We were not provided with an exact reference value for the age of the ZPPR plates but were told that they are “about 33-years old.” The histogram on the left indicates a rather symmetrical distribution of results about the mean age, as illustrated with the superimposed Gaussian, with the exception of several values above 37 years. The plot on the right shows the measured values as they occurred in chronological sequence. The values that measured above 37 years occurred late in the test sequence.

Less precise results were obtained for the other unclassified item, 1.6 kilograms of PuO_2. The reference value for the age of this item was also imprecise, “about eleven years.” The average of our 52 measurements was 12.7 ± 1.2 years. These measurements are considerably less precise than the ZPPR-plate measurements, exhibiting a more noticeable trend toward high values during the latter part of the test period. These rising results were coincident with the onset of drifting in the detector energy calibration, which we suspect accounts for the apparent outliers.

Because a configuration lock was placed on the measurement system for security purposes late in the testing period, we were unable to change out equipment to pursue this hypothesis. Given the imprecision

of the reference age values, the observed instability in our detector at the end of the testing period, and the goal of demonstrating that classified measurement information can be protected, we are not displeased with these results. Nevertheless, a more rigorous set of measurements needs to be carried out to more clearly delineate the expected precision and accuracy of the Pu-300 method.

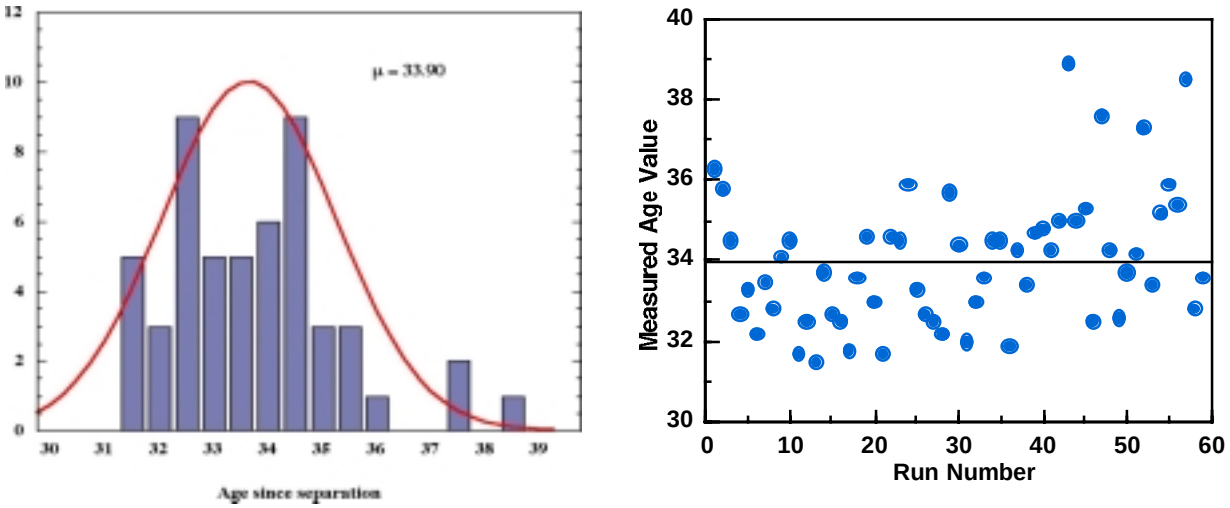

Figure 2. Results of a Pu-300 age measurement of the ZPPR plates. The plot on the left shows the results in the form of a histogram. For reference purposes, we show a Gaussian distribution with the same mean and standard deviation as estimated from the measured data. The plot on the right displays the measurement results in chronological order.

Pu-600—Presence of plutonium oxide

The Pu-600 method was developed in 1994 and has matured incrementally since that time. Pu-600 is also intended to obtain results of adequate accuracy and precision from hundreds of grams of material after acquiring data for 15 minutes. Plutonium emissions in the 630–670-keV region are much weaker than those in the 330–350-keV region used by Pu-300, and the Pu-600 precision was noticeably degraded by the sparse data collected from the ZPPR plates. The average result for the 52 measurements of the isotopic ratio of ^{240}Pu to ^{239}Pu for the ZPPR plates was 0.134 ± 0.014 (or ± 10.5%). The reference value for the ratio was 0.136. The 1.6-kilogram can of PuO_2 was said to contain "about 6% ^{240}Pu." The ^{240}Pu/^{239}Pu result for our 52 measurements was 0.0605 ± 0.0033 (or ± 5.5%, our expected precision for large items with the Pu-600 method).

The results for the ZPPR plates are plotted in Fig. 3. The histogram on the left indicates a well-behaved distribution of results about the mean isotopic ratio. The plot on the right shows the measured values as they occurred in chronological sequence and, unlike the age results, appear to be stable throughout the test period. Similarly well-behaved results were obtained for the PuO_2 item.

Why were these measurements so well behaved when the age measurements were not? We believe these results reinforce the hypothesis that drifts in energy calibration caused the upward creep of Pu-300 age results as the testing period progressed. Because the Pu-300 analysis code must resolve two extremely tight doublets, this code is quite sensitive to small errors in the local energy calibration. On the other hand, by comparison, the peaks in the 630–670 keV region are relatively well spaced, making the Pu-600 analysis code more forgiving of small errors in energy calibration. More careful experiments and a parameter sensitivity study need to be done to verify these assertions.

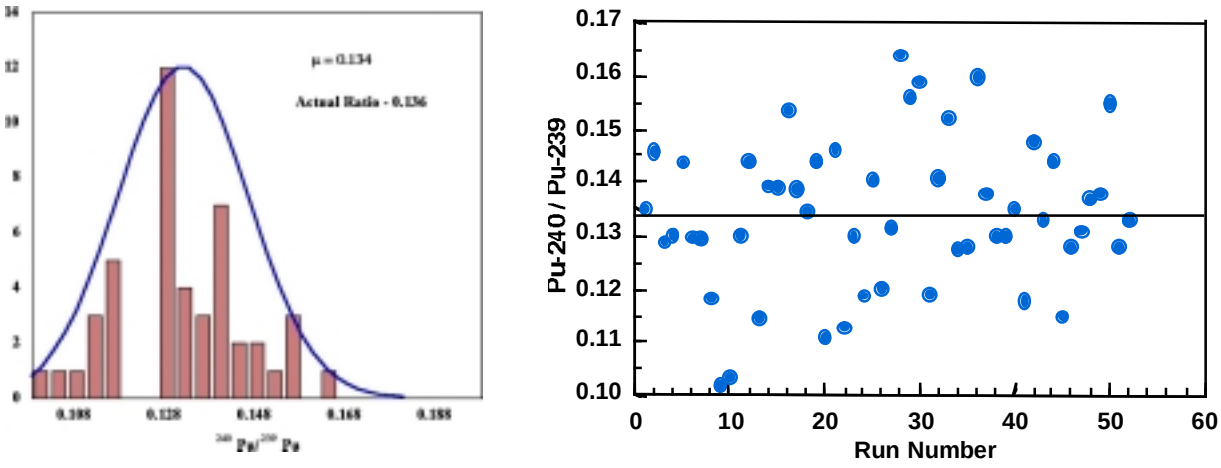

Figure 3. Results of Pu-600 measurements for the ZPPR plates. The plot on the left shows the results in the form of a histogram. For reference purposes, we show a Gaussian distribution with the same mean and standard deviation as estimated from the measured data. The plot on the right displays the measurement results in chronological order.

Pu-900—Absence of plutonium oxide

The absence of plutonium oxide attribute is a surrogate for the truly desired attribute, the presence of plutonium metal. The consensus among several national laboratories is that determining a gamma-ray attribute for the presence of plutonium metal in a sealed container is not technically feasible. Instead, noting that the most probable alternate material form would be plutonium oxide, and that detection of the presence of oxygen seemed possible, it was decided to substitute an absence of PuO_2 attribute.

The initial focus was on a gamma-ray photopeak at 870.7 keV that is absent when the sample is a metal. The 870.7-keV gamma ray is emitted during de-excitation of the first excited state of ^{17}O. It was initially thought the mechanism producing this excited state was due to alpha particles from the decay of plutonium interacting with oxygen by coulomb excitation, an inelastic process, $^{17}O(\alpha,\alpha')$. It was observed by Pacific Northwest National Laboratory (PNNL) that another mechanism is possible—alpha particles reacting with nitrogen impurities in the oxide, $^{14}N(\alpha,p)$. Subsequent work at PNNL[2] and our Laboratory[3] involving PuO_2 samples of varying degrees of chemical purification suggest that the latter mechanism is dominant. Nevertheless, the presence of the 870.7-keV peak unequivocally indicates nonmetallic plutonium. Because the PuO_2 sample used in the FMTTD exhibited a strong 870.7-keV peak, it was decided, due to time pressure, to use this indicator for the FMTTD.

Meanwhile, measurements at our Laboratory demonstrated a possible alternative, the 2,438.0- and 2,788.8-keV peaks from the $^{18}O(\alpha,n)^{21}Ne$ reaction. Unlike the 870.7-keV peak, these ^{21}Ne gamma rays appear to be unambiguously due to the presence of oxygen. These gamma rays, however, are weakly emitted, leading to questions of their practicality, due to length of measurement time, in a realistic transparency regime. The issues surrounding measurement of the presence of PuO_2 by gamma-ray spectrometry are still under investigation.

Performance at the FMTTD

The system performed flawlessly during the demonstration. All measurements functioned normally, the system performed as it should, and determined the correct attributes.

Conclusion

We have described three methods we developed to determine plutonium attributes without revealing classified information. We concentrated on the results of test measurements made with these methods in preparation for the FMTTD. The newest and most challenging of these methods is Pu-300 which determines the age of plutonium in a thick storage container. We described difficulties that have to be overcome in their implementation and problems that need to be addressed before these methods achieves the robustness required for use in an arms control transparency regime.

Reference

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